

Ring vortex destabilization in supersaturated ^3He - ^4He liquid mixtures at low temperatures

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Abstract

The effect of ring vortices on the destabilization of supersaturated liquid helium mixtures at very low temperatures is investigated as a function of pressure. We have found that large ring vortices trigger the segregation of ^3He at smaller values of the concentration than small ring vortices. Our calculations indicate that the existence of ring vortices in the mixture is a possible mechanism to understand the rather small degree of critical supersaturation found in the experiments.

64.60.My, 67.40.Vs, 67.57.Fg, 67.60.-g

I. INTRODUCTION

The study of supersaturated liquid helium mixtures at very low temperatures (T) has received a renewed experimental interest [1–5]. The main goals of these experiments have been, on the one hand, to determine the crossover from the classical thermal regime, to the quantum regime in the nucleation process of the pure ^3He phase from the ^3He - ^4He mixture, and on the other hand, to study the mixture in the metastable supersaturated state, whose existence has been established long ago [6,7].

One of the more interesting observations of recent experiments is the rather small degree of supersaturation achieved in them, of the order of 1 % in [2,4,5] (see also [8]), and below 0.5 % in [1,3,9]. Similar supersaturations were found in [6,7], but at that time no systematic studies of the metastable phase were undertaken.

An extrapolation of the measured ^3He chemical potential excess $\Delta\mu_3$ along the coexistence line yielded $\partial\Delta\mu_3/\partial x \geq 0$ up to values of the ^3He concentration $x > 16$ % [10], indicating that the mixture could be in the metastable, homogeneous phase up to that concentration. That belief seemed further sustained by the calculations of Lifshitz *et al* in their classical paper about the kinetics of the nucleation process in liquid helium mixtures [11], and by microscopic [12] and density functional [13] estimates of the spinodal line in the $P-x$ plane at zero temperature.

Clearly, there is nothing *a priori* contradictory between the results of [12,13] and the small critical supersaturation degree Δx_{cr} found experimentally. Likely, the segregation of ^3He from the mixture occurs before reaching the spinodal line. What is really intriguing is that nucleation calculations [4,11,14], which incorporate the basic physics to consider ^3He droplets as nucleation seeds for phase separation, have yielded results that are around one order of magnitude larger than experiment. Other calculations have put the emphasis in the diffusion mechanism of the quantum decay of the metastable mixture and in the quantum-to-thermal crossover temperature T^* [15], taking as an input the experimental Δx_{cr} . Yet, T^* comes out a factor of four larger than experiment.

It is then quite natural to look for another physical mechanism as a possible explanation for the disagreement. One such mechanism is heterogeneous nucleation. However, the system is extremely pure, and the experimental cells are covered with ^4He , avoiding nucleation on the walls. Since at the low temperatures involved in these experiments, the solubility of ^3He into ^4He is limited and ^4He is still superfluid, one is thus left with an obvious candidate, namely heterogeneous nucleation on vortices whose core is filled with ^3He [16]. This possibility was already mentioned in [5], independently put forward and worked out in same detail in [14], and further developed in [17], as well as in [18].

The basis of the model employed in [14] is that a ^4He vortex line described by a hollow core model (HCM) is gradually filled with ^3He as x increases. In the metastable region, these lines become unstable above a certain Δx value, and the resulting Δx_{cr} turns out to be in the right order of magnitude (around 1.5 %), especially as compared with the experimental results of [2,4]. The HCM results have been confirmed by a more elaborated density functional calculation [17].

The purpose of the present work is to complete the above scenario, studying the effect

that a more realistic vortex geometry, namely a ring vortex, may have on the quantitative results, and also to compare them with the recent experimental results of [8]. As in previous works [14,17,18], the presence of vortices is taken for granted. Indeed, it has been recognized for a long time that ‘any container of superfluid ^4He , treated in conventional fashion, will be permeated *ab initio* by numerous quantized vortices stabilized by surface pinning’ [19]. The interesting problem of vorticity nucleation in liquid ^4He , and the effect of ^3He impurities on it has been addressed in detail in the past (see for example [20–24]).

This work is organized as follows. We present in Section II the model we have used to describe the evolution of a ring vortex as a function of pressure and ^3He concentration. The numerical results are collected in Section III. Although our main interest is in the metastable region in the pressure-concentration ($P - x$) plane at very low temperatures, we have also paid some attention to the stable region, in view that previous studies have not dealt with appreciable ^3He concentrations. A discussion is presented in Section IV, and some technical details are given in an Appendix.

II. RING VORTEX ENERGETICS

To carry out the program we have outlined in the introduction, we first determine the kinetic energy of the ring vortex (RV), for which we assume a hollow core model, i.e., the ^4He density is zero within the vortex core, and equal to the bulk value at the given pressure elsewhere. Following [25,26], we consider the liquid to be incompressible ($\nabla \cdot \vec{v} \approx 0$), so that one can introduce a velocity vector potential $\vec{A}(\vec{r})$ such that $\vec{v} = \nabla \times \vec{A}$. The vector potential is fixed if the vorticity $\vec{\omega} = \nabla \times \vec{v}$ is specified:

$$\vec{\omega} = \nabla \times (\nabla \times \vec{A}) . \quad (1)$$

We make use of the line-source approximation for the vorticity. For a ring vortex with radius of curvature R , circulation number $n = 1, 2, \dots$, and quantum circulation $k_0 = n h/m_4$ located on the $z = 0$ plane, it means that

$$\vec{\omega}(\vec{r}) = k_0 \delta(r - R) \delta(z) \hat{\phi} , \quad (2)$$

where (r, z, ϕ) are the cylindrical coordinates and $\hat{\phi} = (-\sin\phi, \cos\phi, 0)$ is a unit vector. Unless explicitly indicated, we shall be considering $n = 1$ as circulation number. It is straightforward to show that the integral solution of Eq. (1) reads $\vec{A}(\vec{r}) = A_0(r, z)\hat{\phi}$, with

$$A_0(r, z) = \frac{k_0}{4\pi} R \int_0^{2\pi} \frac{\cos\phi' d\phi'}{\sqrt{r^2 + R^2 - 2rR\cos\phi' + z^2}} . \quad (3)$$

The velocity of the superfluid outside the RV can be obtained as

$$\begin{aligned} v_r &= -\frac{\partial A_0}{\partial z} \\ v_z &= \frac{1}{r} \frac{\partial}{\partial r} (r A_0) , \end{aligned} \quad (4)$$

and the kinetic energy of the superfluid is evaluated as an integral over the core surface \mathcal{S} of the ring vortex:

$$E_{kin} = -\frac{m_4}{2}\rho_4 \int_{\mathcal{S}} A_0(r, z) (v_z \hat{r} - v_r \hat{z}) \cdot \hat{n} d\mathcal{S} , \quad (5)$$

where the normal \hat{n} is directed into the ^4He fluid. The boundary condition that no velocity line crosses the vortex core imposes that $\vec{v} \cdot \hat{n} = 0$ and yields

$$\hat{n} = -\frac{v_z}{|\vec{v}|} \hat{r} + \frac{v_r}{|\vec{v}|} \hat{z} \quad (6)$$

as well as the differential equation obeyed by the velocity lines:

$$\frac{dr}{dz} = \frac{v_r}{v_z} . \quad (7)$$

Using Eq. (4), it is easy to verify that solving Eq. (7) to obtain the velocity lines is equivalent to solving the algebraic equation

$$r A_0(r, z) = \text{Constant} . \quad (8)$$

We have specified the cross section profile of the vortex core, compatible with the shape of the velocity lines, introducing a length a such that $(R - a)$ is the closest distance of the core surface to the origin of coordinates:

$$(R - a) A_0(R - a, 0) = C_a . \quad (9)$$

That fixes the constant in Eq. (8) and permits one to obtain the core profile. In the case of large ring vortices with $R \gg a$, for which a circular torus is a good approximation, the length a represents the radius of the circular cross section of the core. We have numerically checked that if $a/R \ll 1$, A_0 at the surface of the core, and the kinetic energy E_{kin} are indeed those of the hollow RV given for example in [25]:

$$A_0 = \frac{k_0}{2\pi} \left[\ln \frac{8R}{a} - 2 \right] \quad (10)$$

$$E_{kin} = \frac{m_4}{2} \rho_4 k_0^2 R \left[\ln \frac{8R}{a} - 2 \right] .$$

We give in the Appendix the general expressions of A_0 , v_r and v_z which are needed to evaluate the kinetic energy, Eq. (5).

Figure 1 shows several cross sections of the core for $R = 30 \text{ \AA}$ and $a = 10, 15$ and 20 \AA , respectively. One can see that these cross sections are very non-circular, especially when the ratio a/R is large.

It is now a simple task to write the energy gain caused by the presence of a ring vortex in the homogeneous mixture at given P . As x increases, the ^3He atoms located at the surface of the vortex migrate to the interior of the hollow core. Within the HCM, one has [14]

$$E_v(R, a) = \sigma \mathcal{S} - \Delta \mu_3 \rho_3 \mathcal{V} + E_{kin} , \quad (11)$$

where \mathcal{V} is the volume of the core, ρ_3 the ^3He atom density inside the vortex core, σ the surface tension of the mixture-pure ^3He interface, and $\Delta\mu_3$ is the ^3He chemical potential excess, i.e., the difference between the chemical potential of ^3He in the metastable mixture, and in pure ^3He . Consequently, $\Delta\mu_3$ has a sign. It is negative when the mixture is stable, and positive when it is not.

The reliability of eq. (11) to describe ^4He vortices filled with ^3He has been checked in [17] for the case of vortex lines using a density functional approach which takes into account the surface diffuseness of both liquids at the interface, and the kinetic energy of the ^4He atoms that penetrate into the vortex core. Altogether, both effects have been found to be small, the reason being that the core radii of critical vortices are large, of the order of 10 Å, and consequently, the sharp surface approximation embodied in the HCM works well. For example, at zero T and P , the HCM yields $\Delta x_{cr} \sim 1.6\%$ [14], whereas the density functional theory yields $\Delta x_{cr} \sim 1.8\%$ [17].

The temperatures of interest here are below 150 mK ([3,4,8]). Bearing in mind that we are looking for a sizeable decrease in the value of Δx_{cr} with respect to that of [11], and that thermal effects are not expected to play an important role because of the small T -dependence of σ [27], $\Delta\mu_3$ [10] and ρ_3 [28] in that temperature domain, we shall carry out a $T = 0$ calculation.

At given P and ^3He concentration $x \equiv \rho_3/(\rho_3 + \rho_4)$, where ρ_4 is the ^4He atom density, the thermodynamical characteristics of the liquid have been described by a density functional that among other properties, reproduces accurately the maximum solubility line and surface tension of the interface as a function of P [29]. We give in Fig. 2 $\Delta\mu_3$ as a function of x for $P = 0, 5$ and 10 atm. The $\Delta\mu_3$ curves intersect the x-axis at a concentration $x_s(P)$ that corresponds to the maximum solubility of ^3He into ^4He for the given P . The slopes $\partial(\Delta\mu_3)/\partial x|_{x_s}$ will be used in the next section to determine the critical supersaturation of the mixture when ring vortices are present, transforming the $\Delta\mu_3$ values used in Eq. (11) into Δx values as

$$\Delta x \approx \Delta\mu_3 \left[\frac{\partial\Delta\mu_3}{\partial x} \Big|_{x_s} \right]^{-1}. \quad (12)$$

This is justified in view of the smallness of Δx . We have obtained $\partial\Delta\mu_3/\partial x|_{x_s} = 2.49, 1.97$ and 1.85 K, and $x_s = 6.40, 9.01$ and 9.58 % for $P = 0, 5$ and 10 atm, respectively.

III. RESULTS

A. Stable phase-diagram region

We have first considered the region in the $P - x$ plane where the homogeneous mixture is stable, i.e., $P \geq 0$ and $\Delta x \equiv x - x_s < 0$, and have minimized the energy Eq. (11) of a RV of radius of curvature R with respect to a . The value of R is considered as an intrinsic characteristic of the vortex that depends on the process of formation, whereas the a value may change with changing x , and its optimum value for given x has to be found through minimization.

As expected from our previous works on vortex lines [14,17], in this case there is only one extremum of $E_v(R, a)$ corresponding to a stable ring vortex. The energy of the equilibrium

configuration and the effective radius of the vortex core a_0 , defined as $\pi a_0^2 \equiv \mathcal{S}$, are drawn in Figs. 3 and 4 as functions of R for $P = 0$ and 10 atm, and $\Delta x = -1, -3$ and -5 %.

B. Metastable phase-diagram region

When Δx is positive, $E_v(R, a)$ at fixed R has in general two extrema, one maximum and one minimum: the ring vortex is no longer stable and may trigger the demixing process of ^3He if it overcomes the barrier whose height ΔE_v is the energy difference between both extrema. It may happen either by thermal or by quantum fluctuations. One such energy barrier is drawn in Fig. 5 for an $R = 100$ Å ring vortex at $P = 0$ and $\Delta x = 1$ %. We show in Fig. 6 the barrier heights as a function of Δx , for different R and P values.

The barrier height decreases with increasing Δx , and there is a critical value Δx_{cr} at which it disappears. This defines a critical ring vortex that depends on the values of R and P , and may freely expand since it is barrierless.

We display in Fig. 7 the effective radius a_0 of the critical vortex as a function of R for $P = 0, 5$ and 10 atm. We see that in contradistinction with the stable case, the core radii are large, and as in the vortex line case, we expect the sharp surface approximation to work well. The critical configurations corresponding to $R = 30$ Å and $P = 0, 5$ and 10 atm are shown in Fig. 8. The associated Δx_{cr} values are 2.75, 1.82 and 1.37 %, respectively. For $R = 100$ Å, these values are 1.67, 1.04 and 0.76 %.

From the previous discussion, it is clear that one can define, for a given R -vortex, a kind of ring vortex spinodal line in the $P - x$ plane, lying in the metastable region, beyond which a liquid helium mixture hosting R -vortices is absolutely unstable. Such spinodal lines can be drawn combining the appropriated $(P, \Delta x)$ values from Fig. 6. We have represented in Fig 9 Δx_{cr} as a function of R for $P = 0, 5$ and 10 atm. From this figure one can see that fixed P , large ring vortices have associated smaller values of Δx_{cr} . It means that large ring vortices trigger the segregation of ^3He at smaller supersaturations than small ring vortices.

IV. DISCUSSION

Very recently, Chavogets et al [8] have carried out a systematic investigation of nucleation in superfluid ^3He - ^4He solutions at $T = 125$ mK as a function of P , which is especially well suited for a detailed comparison with the present calculations since the temperature is kept constant at a value of 125 ± 5 mK. However, these authors stress that it cannot be excluded that the experimental results are influenced by the way the phase transition is initiated, since different methods are used to attain the initial metastable state.

We show in Fig. 10 the experimental results of [8] for Δx_{cr} (dots) and the spinodal values (solid lines) corresponding to ring vortices of $R = 100, 200$ and 500 Å. For $R = 30$ Å the Δx_{cr} values go from ~ 2.7 % at $P = 0$ to ~ 1.5 % at $P = 10$ atm. The calculated Δx_{cr} 's tend to decrease with P , but still are a factor of two larger than the experimental ones even for the most favorable case of large ring vortices. However, the improvement over nucleation calculations that do not consider the possible existence of vortices in the mixture is apparent [2,8,11].

The major discrepancy appears at pressures close to zero atm. Chagovets et al [8] have found that Δx_{cr} has a maximum of 1.5 % around $P = 0.1$ atm, and that Δx_{cr} drops to ~ 0.7 % around $P = 0.02$ atm. We have not found this behavior. However, we would like to recall that at pressures close to zero, the emergence of a free liquid-vapor interface may help nucleate the new phase from the ^3He -enriched surface layer [4]. That might explain the small Δx_{cr} found at low pressures, although the authors of [8] have discarded this possibility [30].

The agreement between theory and experiment can be dramatically improved considering the existence of ring vortices with circulation number $n > 1$. These vortices are rare, since creating an R -vortex with $n = 2$ costs more energy than creating two R -vortices with $n = 1$. This is the reason why in classical experiments where vortices are nucleated and dragged by moving ions only $n = 1$ vortices are detected [31]. Still, it is interesting to look at their effect on Δx_{cr} . Roughly, one should expect that large RV with $n = 2$ originate Δx_{cr} four times smaller than vortices with $n = 1$ (see for example the discussion after Eq. (4) of [14]), thus bringing theory closer to experiment. This is indeed the case, as it can be seen from the dashed lines in Fig. 10 which represent the spinodal values corresponding to ring vortices with $n = 2$ and $R = 30, 50, 75$, and 100 Å.

The above comparisons have involved the calculated spinodal Δx_{cr} values, which are an upper bound to those one would obtain considering thermal activation over the energy barrier (quantum tunneling through it is only effective for temperatures below a few tens of mK, see [3]). We have checked that, although possible, thermal activation plays a minor role. Let us consider, as an example, the case of the $R = 50$, $n = 2$ ring vortex at $T = 125$ mK, $P = 10$ atm, for which the spinodal value is $\Delta x_{cr} = 0.56$ %. Within classical nucleation theory, the decay rate Γ , i.e., the number of RV ‘passing’ over the barrier per unit time and volume due to thermal fluctuations is given by

$$\Gamma = \Gamma_0 \exp(-\Delta E_v/kT) , \quad (13)$$

where the prefactor Γ_0 can be roughly estimated as

$$\Gamma_0 = \frac{kT}{hR^3} . \quad (14)$$

Solving the equation $1 = V_{exp} t_{exp} \Gamma$, taking as experimental volume V_{exp} and time t_{exp} those of [8], and relating the ΔE_v value to Δx , one gets $\Delta x_{cr} = 0.54$ % for $\Delta E_v = 10$ K, and $\Delta x_{cr} = 0.55$ % for $\Delta E_v = 8$ K, which are very close to the spinodal values. A similar conclusion was drawn for vortex lines in [14].

Quantum decay is effective only below a crossover temperature $T^* \sim 10$ mK [3]. None of the calculations carried out so far is able to reproduce Δx_{cr} and T^* simultaneously, and the problem is still open. In the case of vortex lines, T^* has been calculated to be ~ 0.1 mK in [18]. Using a functional integral method similar to that of [32], we have obtained $T^* \sim 1$ mK. The smallness of this value stems from the fact that in the calculations, the decay process is always found to take place very close to the spinodal line.

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APPENDIX:

In this Appendix we give the expressions and method we have used to numerically obtain the velocity lines. From Eqs. (3-4) one has:

$$v_r(r, z) = \frac{k_0 R z}{\pi [(r+R)^2 + z^2]^{3/2}} \int_0^{\pi/2} \frac{2 \cos^2 \phi - 1}{(1 - s \cos^2 \phi)^{3/2}} d\phi, \quad (\text{A1})$$

where we have defined $s \equiv 4rR / [(r+R)^2 + z^2]$. The integral Eq. (A1) is written in terms of hypergeometric functions [33] yielding:

$$v_r(r, z) = \frac{k_0 R z}{2 [(r+R)^2 + z^2]^{3/2}} [F(3/2, 3/2; 2; s) - F(1/2, 3/2; 1; s)]. \quad (\text{A2})$$

Similarly,

$$\begin{aligned} v_z(r, z) = & \frac{k_0 R}{2 r [(r+R)^2 + z^2]^{1/2}} [F(3/2, 1/2; 2; s) - F(1/2, 1/2; 1; s)] \\ & + \frac{k_0 R}{2 [(r+R)^2 + z^2]^{3/2}} [(r+R) F(1/2, 3/2; 1; s) - (r+2R) F(3/2, 3/2; 2; s)] \\ & + \frac{3R}{2} F(5/2, 3/2; 3; s) \end{aligned} \quad (\text{A3})$$

and

$$A_0(r, z) = \frac{k_0 R}{2 [(r+R)^2 + z^2]^{1/2}} [F(3/2, 1/2; 2; s) - F(1/2, 1/2; 1; s)]. \quad (\text{A4})$$

The hypergeometric functions entering Eqs. (A2-A4) can be written in terms of the complete elliptic integrals \mathbf{E} and \mathbf{K} [33] as follows:

$$F(1/2, 1/2; 1; s) = \frac{2}{\pi} \mathbf{K}(\sqrt{s}) \quad (\text{A5})$$

$$F(1/2, 3/2; 1; s) = F(3/2, 1/2; 1; s) = \frac{2 \mathbf{E}(\sqrt{s})}{\pi (1-s)} \quad (\text{A6})$$

$$F(3/2, 1/2; 2; s) = F(1/2, 3/2; 2; s) = \frac{4}{\pi s} [\mathbf{K}(\sqrt{s}) - \mathbf{E}(\sqrt{s})] \quad (\text{A7})$$

$$F(3/2, 3/2; 2; s) = \frac{4}{\pi s} \left[\frac{\mathbf{E}(\sqrt{s})}{1-s} - \mathbf{K}(\sqrt{s}) \right] \quad (\text{A8})$$

$$F(5/2, 3/2; 3; s) = \frac{16}{3\pi s^2} \left[\frac{2-s}{1-s} \mathbf{E}(\sqrt{s}) - 2\mathbf{K}(\sqrt{s}) \right]. \quad (\text{A9})$$

To evaluate \mathbf{E} and \mathbf{K} we have used accurate polynomial approximations [34].

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FIGURES

FIG. 1. Core cross sections corresponding to ring vortices of radius of curvature $R = 30 \text{ \AA}$ and a -values of 10, 15 and 20 \AA . The vorticity line is indicated by a dot.

FIG. 2. ^3He chemical potential excess $[\mu_3(x) - \mu_3(\text{pure})]$ (K) as a function of x (%) for $P = 0, 5$ and 10 atm .

FIG. 3. Bottom panel: Ring vortex energy E_v (K) as a function of R (\AA) for $P = 0$. Top panel: same as bottom panel for $P = 10 \text{ atm}$. Both panels correspond to stable vortices, and the curves represent the results for $\Delta x = -1 \%$ (solid line), -3% (dotted line) and -5% (dashed line).

FIG. 4. Bottom panel: Effective radius a_0 (\AA) of the vortex cross section as a function of R (\AA) for $P = 0$. Top panel: same as bottom panel for $P = 10 \text{ atm}$. Both panels correspond to stable vortices, and the curves represent the results for $\Delta x = -1 \%$ (solid line), -3% (dotted line) and -5% (dashed line).

FIG. 5. Ring vortex energy E_v (K) as a function of a (\AA) for an $R = 100 \text{ \AA}$ ring vortex at $P = 0$ and $\Delta x = 1 \%$.

FIG. 6. Barrier heights ΔE_v (K) as a function of Δx for $P = 0, 5$ and 10 atm . From top to bottom, the panels correspond to $R = 10, 30$ and 50 \AA .

FIG. 7. Effective radius a_0 (\AA) of the critical vortex as a function of R (\AA) for $P = 0, 5$ and 10 atm .

FIG. 8. Core cross sections of critical ring vortices corresponding to $R = 30 \text{ \AA}$ and $P = 0, 5$ and 10 atm .

FIG. 9. Critical supersaturation Δx_{cr} (%) as a function of R for $P = 0, 5$ and 10 atm .

FIG. 10. Critical supersaturation Δx_{cr} (%) as a function of P (atm). The dots are the experimental values of Ref. [8]. From top to bottom, the solid lines represent the spinodal values for ring vortices with $R = 100, 200$ and 500 \AA and circulation number $n = 1$, and the dashed lines represent the spinodal values for ring vortices with $R = 30, 50, 75$ and 100 \AA and circulation number $n = 2$.



















